Spectrum and polarization of helium doubly excited triplet states

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The beam-foil spectrum of helium from 2050 to 3600 Å has been acquired at 160-keV incident ion energy using a multiple-scan, computerized system. Eleven doubly-excited-state transitions, including two not previously observed, have been assigned, and the agreement between experimental and theoretical energy levels is excellent. The Stokes parameters as a function of foil-tilt angle have been measured for the two strongest transitions (2578 and 3013 Å) from 0° to 80° in 10° increments and for two weaker transitions (2562 and 2818 Å) at 0°, 20°, 40°, and 60°. The polarization patterns are quite different from those of the helium singlets and include the first measurement of negative M/I (a Stokes parameter) at 0° foil tilt for helium at this energy. The lifetime of the 2578-Å transition has been investigated in detail. The present result, 0.109 ± 0.004 ns, agrees with previous measurements and yields an upper limit to the autoionization width of the $2p3p\ ^3D$ levels of 6×10^{-6} eV.

I. INTRODUCTION

One of the interesting aspects of the beam-foil light source is that many doubly excited states in helium (He**) and other elements are easily observable in emission. Although all He I doubly excited states lie above the first ionization limit, selection rules prevent autoionization for many of the levels, which then decay by radiation. Furthermore, many of the states which can autoionize do so at a sufficiently slow rate that radiative decay becomes competitive. We have previously published the results of some polarization measurements for singly excited He I singlet transitions.[1] In this present report we discuss our more recent investigation of He I transitions from doubly excited triplet states in the spectral region from 2050 to 3600 Å.

II. EXPERIMENTAL PROCEDURES

The spectral, polarization, and lifetime measurements presented here were all made using the computer-controlled facility described previously.[1] For the spectral recordings, the incident energy of the helium ions was maintained at 154 ± 4 keV, yielding an energy after the 5-μg/cm^2 carbon foils of 147 ± 5 keV. The beam current used was 6.5 $\pm$ 1.0 μA. The radiation was dispersed using a $\frac{3}{4}$-meter Czerny-Turner spectrometer equipped with a 2400-groove/mm holographic grating. The spectra were recorded with the spectrometer slits set perpendicular to the beam direction and having widths of 0.5 mm, corresponding to an instrumental bandpass of approximately 2.6 Å. Ten successive spectral scans, each recorded in steps of 1 Å using beam-current normalization, were superimposed to produce the final spectra.

The polarization measurements were made by observing the radiation emitted from the region immediately after the foil through a polarimeter equipped with a zero-order retarder (quarter-wave at 3650 Å) followed by a linear polarizer. The Stokes parameters (M/I, C/I, and S/I) and detector geometry are defined in Ref. 1. The retarder was stepped in 10° increments from 0° to 360° using optical normalization, while the linear polarizer was held fixed. A typical set of data for a given foil angle consisted of the sum of five such sweeps for the lines at 2578 Å ($2s2p\ ^3P^o$-$2p3p\ ^3D$) and 3013 Å ($2p^2\ ^3P$-$2p3d\ ^3D^o$), although many more sweeps (typically 15 to 20) were necessary for the much weaker lines at 2562 Å ($2p^2\ ^3P$-$2p4d\ ^3D^o$) and 2818 Å ($2p^2\ ^3P$-$2p3d\ ^3P^o$). This measurement was made at each foil-tilt angle from 0° to 80° in 10° increments. The slits of the spectrometer were tilted to remain parallel to the foil up to 60° tilt (the maximum tilt possible on our spectrometer), so that some averaging along the beam was unavoidable for the 70° and 80° foil-tilt measurements. The phase of the retarder was determined to within ±1° at each of the wavelengths studied, using the procedures described previously.[1] During the polarization measurements, the incident ion energy was held at 163 ± 5 keV, and the beam current at 6.5 ± 1.0 μA over an area perpendicular to the beam direction of 5 mm diameter, defined by an aperture upstream from the foil. By using elongated foils ($\sim 5 \times \sim 15$ mm^2), it was possible to pass the entire incident beam for all tilt angles used, except 80° for which 3 μA passed through the foil.

The lifetime measurements at 2578 Å were conducted using a higher incident ion energy of 270 ± 10 keV to improve the time resolution. A decay curve measurement involved the superposition of typically 25 sweeps. Each sweep consisted of 50 points obtained by moving the foil in steps of 0.05 mm controlled by beam-current normaliza-

tion. The decay was thus followed for ~0.7 ns from the foil. The spectrometer slits were set at 0.25 mm. The averaging length of our arrangement was measured directly from the initial intensity rise at the foil to be ~0.3 mm, corresponding to a time interval of about 80 ps. Energy-loss corrections appropriate to the foil-tilt angles used were made in computing the post-foil beam velocities.

III. SPECTRAL ANALYSIS

The spectrum of helium between 2000 and 2600 Å is shown in Fig. 1. Nine doubly excited transitions have been observed within this region, as well as eight members of the He II $3d\,^2D$-$nf\,^2F^o$ series. The autoionization rates of these doubly excited transitions have been shown to be sufficiently low[2,3] to permit radiative decay to be at least competitive. (Some controversy does still exist concerning the autoionization widths of the $2pnp\,^3D$ levels, as will be discussed later in this report.) Two spectral features, one at 2479.0 ± 1.0 Å and one at 2065.8 ± 1.5 Å, have been observed which we cannot assign to helium. The first is almost certainly the 2478.6-Å transition of C (sputtered from the foil) while the second may be the 2064.0-Å transition of N III (residual gas excitation).

Table I lists the observed transition wavelengths in air and the corresponding upper-state energies (including the wavelength correction to vacuum). For convenient identification we have employed simple spectroscopic notation corresponding to the dominant configuration. The experimental energies are based on values published for the $2s2p\,^3P^o$ (470310 ± 50 cm^{-1}) and $2p^2\,^3P$ (481301.5 ± 1.2 cm^{-1}) terms by Martin[4] and by Tech and Ward,[5] respectively. The theoretical values are from two recent papers.[6,7] The agreement is impressive, particularly as the theoretical values have been obtained *ab initio*. (We have included in Table I two additional transitions outside the spectral region shown in Fig. 1.)

^3P-$^3D^o$. Five transitions of the type $2p^2\,^3P$-$2pnd\,^3D^o$ have been observed (for $n=3$ to 7). Only radiative decay is permitted for members of this series.

$^3P^o$-^3P. Only one transition of this type, $2s2p\,^3P^o$-$2p3p\,^3P$ at 2364 Å, has been observed. Higher members of this series would lie below 2050 Å. Again only the radiative decay mode is allowed.

^3P-$^3P^o$. Three members of the $2p^2\,^3P$-$2pnd\,^3P^o$ series have been observed (for $n=3$ to 5) though all upper states are permitted to autoionize. Lipsky and Conneely[8] have calculated the autoionization widths for many of the $^3P^o$ terms using designations which reflect the strong configuration interaction required to obtain accurate energy levels. The widths for the above terms were found to be quite narrow (of order 10^{-6} eV) which explains our ability to observe radiative emission from

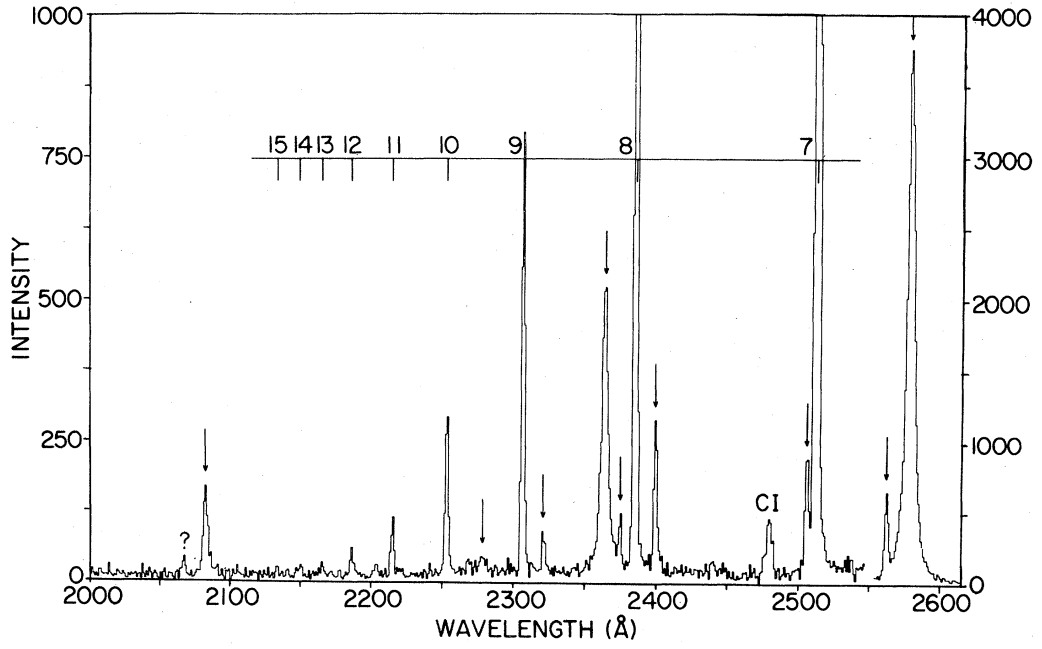

FIG. 1. Beam-foil spectrum of 150-keV He^+ ions incident on a 5-$\mu g/cm^2$ carbon foil. The solid bar indicates members of the He II $3d$-nf series, while the arrows indicate He I doubly excited triplet transitions. Note the change of intensity scale above 2550 Å.

TABLE I. Doubly excited He I transitions observed in this work.

λ (Å)$_{air}$ [a]	Transition	Energy of upper state (eV) This work [a]	Theory [b]	Theory [c]
2081.5(1.0)	$2s2p\,^3P^o$-$2p4p\,^3D$	64.266(7)	64.268	64.274
2278.2(1.0)	$2p^2\,^3P$-$2p7d\,^3D^o$	65.117(3)		65.117 [d]
2321.2(0.5)	$2p^2\,^3P$-$2p6d\,^3D^o$	65.014(2)	65.012	65.013
2364.0(0.5)	$2s2p\,^3P^o$-$2p3p\,^3P$	63.554(6)	63.557	63.585
2375.2(0.5)	$2p^2\,^3P$-$2p5d\,^3P^o$	64.893(2)	64.902	64.894
2399.8(0.5)	$2p^2\,^3P$-$2p5d\,^3D^o$	64.839(2)	64.838	64.839
2505.3(1.0)	$2p^2\,^3P$-$2p4d\,^3P^o$	64.622(3)	64.624	64.626
2561.9(0.5)	$2p^2\,^3p$-$2p4d\,^3D^o$	64.512(2)	64.513	64.516
2577.6(0.5)	$2s2p\,^3P^o$-$2p3p\,^3D$	63.120(7)	63.125	63.136
2817.6(0.5)	$2p^2\,^3P$-$2p3d\,^3P^o$	64.073(2)	64.076	64.079
3013.0(1.0)	$2p^2\,^3P$-$2p3d\,^3D^o$	63.788(3)	63.789	63.795

[a] Uncertainties shown in parentheses.
[b] *Ab initio* calculation by Calloway (Ref. 6).
[c] *Ab initio* calculation by Lipsky *et al.* (Ref. 7).
[d] Using an extrapolated value for $n^*=6.929$ (see Ref. 7).

these levels. Our failure to observe a transition near 3470 Å, which has been reported elsewhere,[9] may be explained by the very short lifetime[8] (~0.01 ns) against autoionization calculated for the upper state.

$^3P^o$-^3D. Two transitions of this type were observed belonging to the series $2s2p\,^3P^o$-$2pnp\,^3D$ ($n=3,4$). One of these (at 2578 Å) is the strongest doubly excited transition in the region above 2000 Å. Recently Conneely and Lipsky[2] have included the $^3D^e$ terms in their analysis. Their calculated width (2.14×10^{-4} eV) for the $2p3p\,^3D$ levels is, however, much too broad to allow an observable radiative decay. These widths have also been calculated by several other authors[3,10,11] and a comparison of the various results is shown in Table II, together with the corresponding lifetimes against autoionization. Our observed value for the lifetime of the $2p3p\,^3D$ term is 0.109 ± 0.004 ns, implying an autoionization width not greater than $\sim6\times10^{-6}$ eV. (We discuss this lifetime measurement in more detail later in this report.)

IV. POLARIZATION OBSERVED USING A TILTED FOIL

Only two of the observed doubly-excited-state transitions are sufficiently intense to allow a complete sequence of polarization measurements from 0° to 80° foil-tilt angle. The first is the $2s2p\,^3P^o$-$2p3p\,^3D$ transition at 2578 Å and the second is the $2p^2\,^3P$-$2p3d\,^3D^o$ transition at 3013 Å. The results for these transitions are presented in Fig. 2. The uncertainties shown for the relative Stokes parameters are calculated within the data-analysis routine described elsewhere,[1,12] and are determined ultimately by the counting statistics. The larger error limits at 80° foil tilt thus reflect the weaker signals resulting from the partial obstruction of the beam by the foil aperture at this large angle. (The increased averaging length at 70° and 80° foil tilt mentioned earlier should not significantly affect the relative Stokes parameters unless measurable quantum beat oscillations are present. A direct measurement of the intensity decay curve for the 2578-Å transition to be discussed later in this report failed to reveal any quantum beats at 60° foil tilt, presumably because the beat frequencies were too high, even with a spatial resolution better than that which was used in the measurements shown in Fig. 2.)

The first striking feature is that M/I for both transitions is essentially constant with tilt angle but of opposite sign. The 3013-Å transition is the only one in helium to show a negative value at

TABLE II. Theoretical energies and autoionization widths for the ^3D term at 63.120 eV.

Author	Energy (eV)	Width (Γ) (eV)	Lifetime ($\hbar/\Gamma$) (ns)
Altick and Moore (Ref. 10)	63.157	1.2×10^{-5}	0.05
Bhatia and Temkin (Ref. 3)	63.120	2.7×10^{-6}	0.24
Conneely and Lipsky (Ref. 2)	63.136	2.1×10^{-4}	0.003
Cooper *et al.* (Ref. 11)	63.141	1.4×10^{-6}	0.46

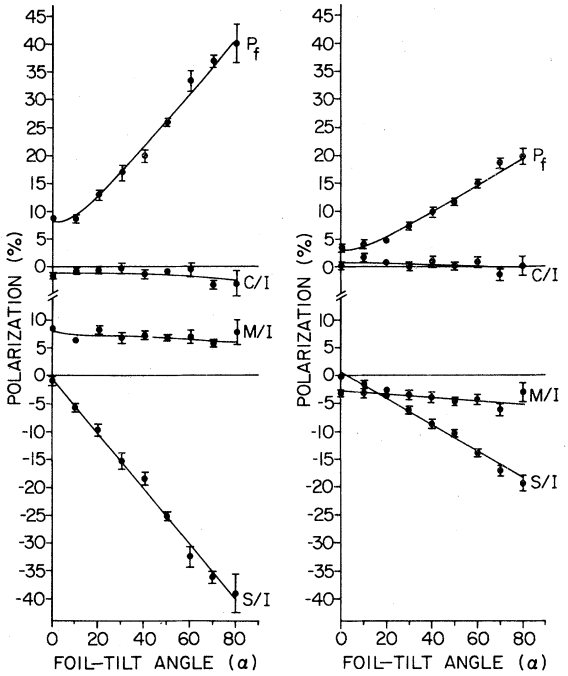

FIG. 2. Polarization of the He** transitions at 2578 Å (on left) and 3013 Å (on right) as a function of foil-tilt angle (α), showing M/I, S/I, C/I, and P_f (total polarization). The error bars have been derived from the computer analyses (see Refs. 1 and 12). The solid lines are smooth curves drawn to aid the eye.

160 keV for M/I at 0° foil tilt. In fact it is one of the few nonhydrogenic transitions of any element to show negative M/I for a perpendicular foil.

S/I for both transitions shows the same trend with foil tilt, this being somewhat intermediate between the behavior of the $^1P^o$ and ^1D curves of neutral helium.[1] S/I reaches a value of nearly −40% at 80° foil tilt for the 2578-Å transition, which is one of the largest polarization fractions ever measured with a tilted foil.

Two much weaker lines have also been studied at 0°, 20°, 40°, and 60°. One is the $2p^2\,^3P$-$2p3d\,^3P^o$ at 2818 Å. All of the polarization values at all four tilt angles are quite small and consistent with their value being zero. The other transition is the $2p^2\,^3P$-$2p4d\,^3D^o$ at 2562 Å. The upper state is simply the next-higher member of the $^3D^o$ series which gave rise to the 3013-Å transition and might then be expected to yield the same polarization. However, this line is in the wing of the 2578-Å line, which is very broad, and hence some fraction of the observed intensity is coming from that line. The polarization values for 2562 Å at each of the four measured tilt angles may be satisfactorily reproduced by admixing 70% of the 3013-Å values with 30% of the 2578-Å values.

There are two complications which exist for these doubly excited transitions which did not occur for the He I singlets considered in our earlier work.[1] First, the lifetimes of the radiating states are all relatively short, and hence it is not practical to attempt polarization measurements at any position other than at or close to maximum intensity immediately after the foil. With a beam energy of 160 keV and a lifetime of 0.1 ns, the light intensity decreases by 65% in 0.3 mm, which is less than the averaging length due to the spectrometer slit widths (0.5 mm) used for the polarization measurements. The second complication arises from the fact that the radiating states are triplets and hence possess fine structure not resolved in our experiment. The presence of quantum beats in the decay curve is therefore to be expected, resulting in the polarization of the emitted radiation being a function of distance along the beam. Without knowing the fine-structure separation it is not possible to determine whether one is measuring some average value of the polarization along the beat or some value close to the initial peak.

Of particular interest is the negative M/I value found at 0° tilt for the $2p^2\,^3P$-$2p3d\,^3D^o$ transition at 3013 Å. For the case of unresolved fine structure, M/I may be related to the alignment parameters of Fano and Macek by the equation[12,13]

$$\frac{M}{I}=\frac{\frac{3}{4}h^{(2)}G^2(t)[A_0^{\mathrm{col}}(0)-A_{2^+}^{\mathrm{col}}(0)]}{1+\frac{1}{4}h^{(2)}G^2(t)[A_0^{\mathrm{col}}(0)+3A_{2^+}^{\mathrm{col}}(0)]}$$

with

$$h^{(k)}(L,L_f)=(-1)^{L-L_f}\begin{Bmatrix}L & L & k\\ 1 & 1 & L_f\end{Bmatrix}\begin{Bmatrix}L & L & k\\ 1 & 1 & L\end{Bmatrix}^{-1}$$

and

$$G^k(t)=\sum_{JJ'}\frac{(2J+1)(2J'+1)}{(2S+1)}\begin{Bmatrix}J & J' & k\\ L & L & S\end{Bmatrix}^2\cos(\omega_{JJ'}t)\,.$$

Here all quantum numbers relate to the upper states except L_f, the orbital angular momentum of the lower state.

One might question whether the negative M/I is a consequence of the excitation mechanism or simply reflects the phase of a partially resolved quantum beat. The latter effect cannot be responsible since M/I can shift sign as a function of distance from the foil, for a ^3P-^3D transition, only if all three possible frequencies in the expression for $G^2(t)$ are nearly in phase and one happens to be viewing the beam at the minimum-amplitude position. Since the experiment averages the light in-

tensity over more than one lifetime, close to the foil, and the phases are known to be zero at the foil, it is not possible for such a position to be resolved. The conclusion then is that the alignment, represented by $A_0^{col}(0)$ for 0° foil tilt, is positive for the 3013-Å transition which is opposite in sign to that of the 2578-Å transition. Extending the argument shows that the alignment of the $2p3d\,^3D^o$ term must be opposite to that of all other measured He I states at 0° foil tilt and at 160-keV incident ion energy.

(The other possible explanation for the negative M/I value for the 3013-Å transition—that it is incorrectly assigned—seems highly unlikely in view of the close agreement with theory for the $2p3d\,^3D^o$ energy, plus the observation of the next four members of the Rydberg series. Furthermore, the relatively narrow width of this line—less than 4 Å in our spectra—compared with the much wider lines at 2364 and 2578 Å is consistent with its assignment as a transition between two states forbidden to autoionize.)

V. LIFETIME MEASUREMENTS AT 2578 Å

As was mentioned in the discussion of Table II, considerable disagreement exists between the values calculated by various authors[2,3,10,11] for the autoionization width of the $2p3p\,^3D$ levels. Previous beam-foil experiments have measured the lifetime of this level as 0.14 ± 0.02 (Ref. 9) and 0.115 ± 0.006 ns (Ref. 14). However, no discussion is given in those reports of the possible effects of the presence of either cascading or quantum beats. Furthermore, because of the relative weakness of this line, the peak counts used in those measurements were rather low ($\lesssim 10^3$). Since our computer-controlled facility permits us to superimpose precisely many successive decay curve sweeps, we were able to improve the counting statistics considerably over the earlier measurements. We now discuss the questions of time resolution, cascading, and quantum beats as they apply to the 2578-Å transition.

From our recent discussions of the relationship between the optical averaging length along the beam and the shortest measurable lifetime,[15,16] we conclude that the arrangement used in the present work should be capable of detecting lifetimes at least as short as ~25 ps. This time resolution capability, together with the relatively high peak counts (~several thousand), should reveal any significant cascading in the decay curves, particularly as there is no reason to expect the severe cascading problems recently discussed in the literature for heavy elements (see Ref. 15). Having tried a variety of fitting functions with the program, TROY,[15] we find that all our 2578-Å decays may be consistently analyzed using a function containing a single exponential term plus an adjustable constant to represent contributions from cascades which are long-lived relative to the time scale of the measurement. [We also found some evidence for a very weak growing in cascade with a lifetime of 0.02 ± 0.02 ns in each decay, presumably from a higher state (or states) able to autoionize and hence having a relatively short life time. This component is not shown in Table III as it is so poorly defined because of its low amplitude. The primary lifetime derived from our measurements was virtually unaffected by the inclusion or omission of this very weak additional short-lived component.]

The presence of quantum beats in a decay curve recorded with a perpendicular foil can be eliminated by inserting a linear polarizer into the optical channel with its axis at 54.7° to the beam direction.[17] We have recorded decay curves for the 2578-Å transition using the four optical configurations shown in Table III. For the measurements made at 60° foil tilt, the beam velocity has been adjusted in the analysis to allow for the increased energy loss at the foil, and also the tilted spectrometer slits were reduced to a width of 0.125 mm to maintain the same spatial resolution. Comparing the results of the first two configurations (at 0°) in Table III tests the effect of the linear polarization, while comparing the second pair (at 60°) tests the effect of the much larger circular polarization present at 60° foil tilt (see Fig. 2). In both cases, the analysis for the data which should reveal the presence of quantum beats does have a somewhat higher χ^2 value, but no effect is apparent on the lifetime derived from the analysis. Fourier transforms of these decay curves failed to show any significant peaks. We conclude that the fine-structure intervals must give rise to beat frequencies which are at least comparable to that corresponding to our averaging length, viz., 12 GHz.

We conclude that the effect of quantum beats on our lifetime measurements for the 2578-Å He I transition must be small. The weighted mean of the values shown in Table III is 0.109 ± 0.004 ns, where the major source of error arises from the uncertainty in the beam velocity. This result is in excellent agreement with that recently reported by Ishii and Tomita,[14] viz, 0.115 ± 0.006 ns, and requires that the autoionization width of the $2p3p\,^3D$ levels cannot be greater than $\sim 6 \times 10^{-6}$ eV. As may be seen from Table II, this is in closest agreement with the value calculated by Bhatia and Temkin.[3] Interestingly enough, their value for the energy of the $2p3p\,^3D$ is also in closest agreement with experiment.

TABLE III. Lifetime results for the 2578-Å He I** transition.

Foil-tilt angle[a]	$\frac{1}{4}\lambda$-Retarder angle[a]	Linear polarizer angle[a]	Lifetime (ns)	χ^2[b]
0°		90°	0.111 ± 0.003[c]	0.964
0°		36°	0.108 ± 0.003[c]	0.801
60°		36°	0.107 ± 0.003[c]	0.773
60°	45°	90°	0.111 ± 0.005[c]	1.332

[a] Angles defined with respect to the vertical (i.e., 90° is parallel to beam).
[b] χ^2 per degree of freedom.
[c] Fitting function required an additional small adjustable constant, presumably to account for long-lived cascades.

As a footnote, we wish to mention the excellent agreement between our measurement of the $2p3p\ ^3D$ lifetime and the results obtained by Berry *et al.*[18] using transitions at 294 Å (0.116 ± 0.020 ns) and 309 Å (0.105 ± 0.015 ns), which are the wavelengths expected for the $1s2p\ ^3P^o$-$2p3p\ ^3D$ and $1s3p\ ^3P^o$-$2p3p\ ^3D$ transitions, respectively. However, possible blending with singlet transitions[9,18] renders the assignment of the measured lifetimes to the $2p3p\ ^3D$ term less certain in these cases.

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